Supplementary Materials

Text S1 Description of ART-2a

The artificial neural network method (ART-2a) has been widely used to analyze the data of single particles measured via aerosol time-of-flight mass spectrometry (ATOFMS) and SPAMS. According to previously published studies (Hopke and Song, 1997; Song et al., 1999; Song et al., 2001), the ART-2a algorithm follows specific steps to obtain classification results for single particles. 1) Initialization: Set the parameters of the ART-2a algorithm, including the vigilance factor (a value between 0 and 1 that controls the classification strictness), the learning rate, and the number of iterations. In this work, these parameters were set as follows: a vigilance factor of 0.75, a learning rate of 0.05, and 20 iterations. 2) Create internal templates: Each recognition unit in the ART-2a network represents an internal template that characterizes a specific class of mass spectra. Initialize these internal templates randomly or with predefined values. 3) Input pattern matching: For each mass spectrum, calculate the matching degree between the input pattern (m/z values and peak areas) and each recognition unit's internal template. The matching degree is typically computed using a similarity measure. 4) Competition and learning: Identify the recognition unit with the highest matching degree as the winner. If the matching degree surpasses the vigilance parameter, assign the input pattern to the corresponding class represented by the winner. Otherwise, create a new recognition unit and update its internal template with the input pattern. 5) Update internal templates: After assigning the input pattern to a class or creating a new recognition unit, update the internal template of the winning recognition unit using the learning rate and the input pattern. This update refines the internal template to better represent its associated class. 6) Repeat iterations: Iterate steps 3) to 5) for all mass spectra until the convergence criteria are met. By repeating these steps, the ART-2a algorithm iteratively adjusts the internal templates to classify the mass spectra into different classes based on their m/z values and peak areas. The resulting classes represent distinct patterns or groups of mass spectra that share similarities in their ion compositions and peak intensities.

Table S1 Particle counts of total single particles and OOM particles, and the number fraction (N_f) of the OOM particles in January, April, August, and October

Month	Total particles	OOM particles	N_f of OOM particles
January	1022779	225044	0.22
April	524005	182287	0.35
August	224669	130933	0.58
October	391724	237323	0.61

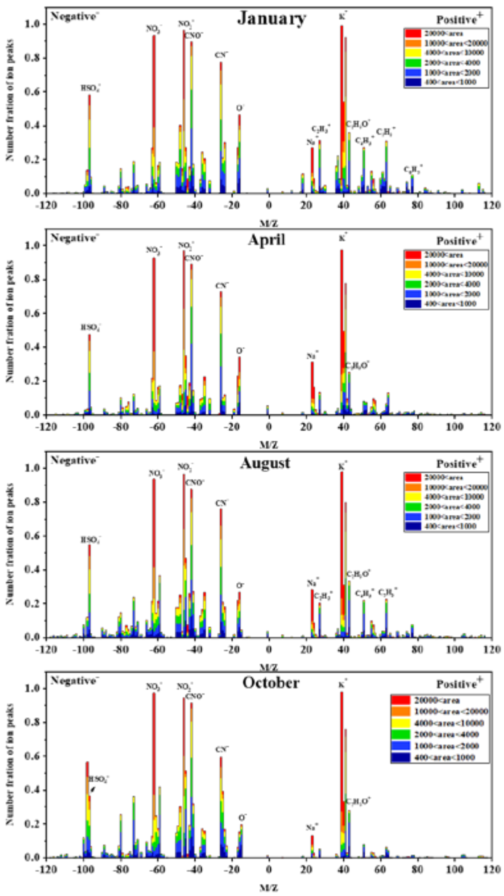

Fig. S1 Mass spectra of the SEC-OOM particles in January, April, August, and October.

References

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